Enabling Rapid Charging in Lithium-Ion Batteries via Integrated Acoustofluidics

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Collaboration with: Ofer Manor, Technion (Israel)

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Overview

- Timeline
  - Start: 2018-07-01
  - End: 2020-06-30
  - Percent complete: 40%
- Budget
  - Total project funding
    - DOE share: $726,242
    - Contractor share: $72,625
  - Funding for FY 2018: $356,226
  - Funding for FY 2019: $370,016
- Barriers addressed
  - **Performance:** Our electrochemistry-agnostic technology enables fast charging >3C with significant improvement (20%) in discharge capacity for Li-ion 2Ah batteries.
  - **Cost, Recycling & Sustainability:** Improve lithium utilization through longer battery lifetime and reduced risk of dendrite and dead Li deposition. Potential selection of electrochemistries previously not possible that would reduce cost and improve sustainability (i.e. Li metal).
- Partners
  - Qualcomm Institute
  - Ofer Manor, Technion (Israel)
  - Ilenia Battiato and Daniel Tartakovsky, Stanford University

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Relevance

- ## Overall Objective
  - Reduce the diffusion-limited Li+ ion depletion layer's thickness in a lithium ion battery using surface acoustic wave-driven mixing flow of the electrolyte during charging.
  - Targeting an energy density of 200 Wh/kg at a 6 C charging rate in a 2 Ah LIB capable of at least 500 cycles and that is able to maintain at least 80% of the initial energy density.
- ## Objectives this period
  - Design, fabricate, and demonstrate a SAW-integrated prismatic 2 Ah LIB capable of 6 C charging rates with 200 Wh/kg energy density and <20% fade in this energy density after 500 cycles. **(1)**
  - Produce theoretical models sufficient for design purposes to serve in integrating SAW technology into prismatic and jelly-roll battery configurations typical in EV applications. **(2)**
  - Produce prototype circuits appropriate for battery cycling and operating SAW device as demonstration of future integration of technology into batteries. **(3)**
- ## Impact
  - Targeting the XFC* goal to **(1,3)** research [and] develop, and gain **(2)** a better fundamental understanding of next generation fast charge battery cells.

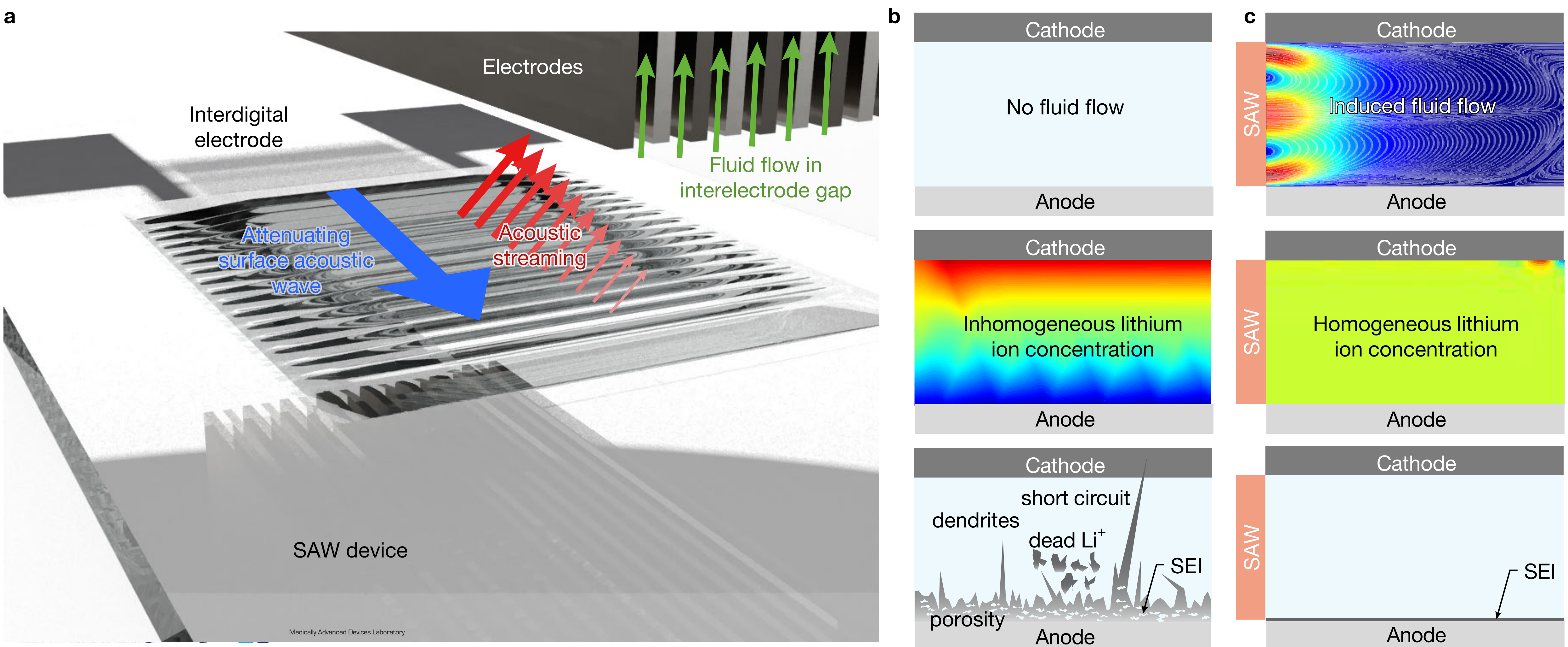
*Batteries for Extreme Fast Charging

Milestones

Milestone	Description	Status	Due
Prismatic Multilayer LIB Constructed	Construction of a prismatic multilayer LIB incorporating SAW appears feasible: at least one design is produced that can be fabricated.	Complete	Sep 2018
Battery Testing Initiated	Battery testing is underway, with 6 or more batteries in test or completed testing.	Complete	Dec 2018
Battery Design Initiated	Initial design of jellyroll SAW LIB is feasible, implying at least one version of the jellyroll design appears to be manufacturable and testable.	Complete	Mar 2019
Analysis and Modeling Initiated	Consistent and complete set of equations to represent turbulent acoustic streaming-driven flow of fluids with dilute species' concentration and diffusion.	Complete	Mar 2019
LIB Design Frozen	Final design freeze for prismatic 2 Ah LIB capable of 6 C charging rates with 200 Wh/kg energy density and <20% fade in this energy density after 500 cycles.	On track	Jun 2019
Go/No Go Decision	*Delivery of 9 prismatic 2 Ah LIB per the specifications defined in task 1.4 to the Department of Energy.*	On track	Jun 2019
Control Circuit Design Complete	Portable circuit design complete, including automatic sequencing of signal generator and battery cycler.	Complete	Sep 2019
Battery Produced	At least one fabricated jellyroll SAW LIB 20700 cell battery produced and suitable for future fast charging (~ 6 C) protocols to >250 cycles.	Future	Sep 2019
Battery Design Report Complete	A compiled report of the quantitative results demonstrating the jellyroll SAW LIB 20700 battery design, with recommendation for final design.	Future	Dec 2019
Report Created	Confirm the performance characteristics of the final design, identify discrepancies, and report small-batch manufacturing and testing results.	Future	Mar 2020
Report Created	Compile and report model of [acoustic streaming-enhanced electrolyte] phenomenon in comparison to experimental results.	Ahead of schedule	Jun 2020
Report Created	Compile and report results of modeling effort in aiding the jellyroll 20700 design and its utility in predicting the acoustic streaming-driven flow in that system.	Future	Jun 2020
End of Project Go/No-Go	*Delivery of 18 jelly roll 2 Ah LIB and 2 circuit board per the Q8 milestone specifications to the Department of Energy.*	Future	Jun 2020

Approach

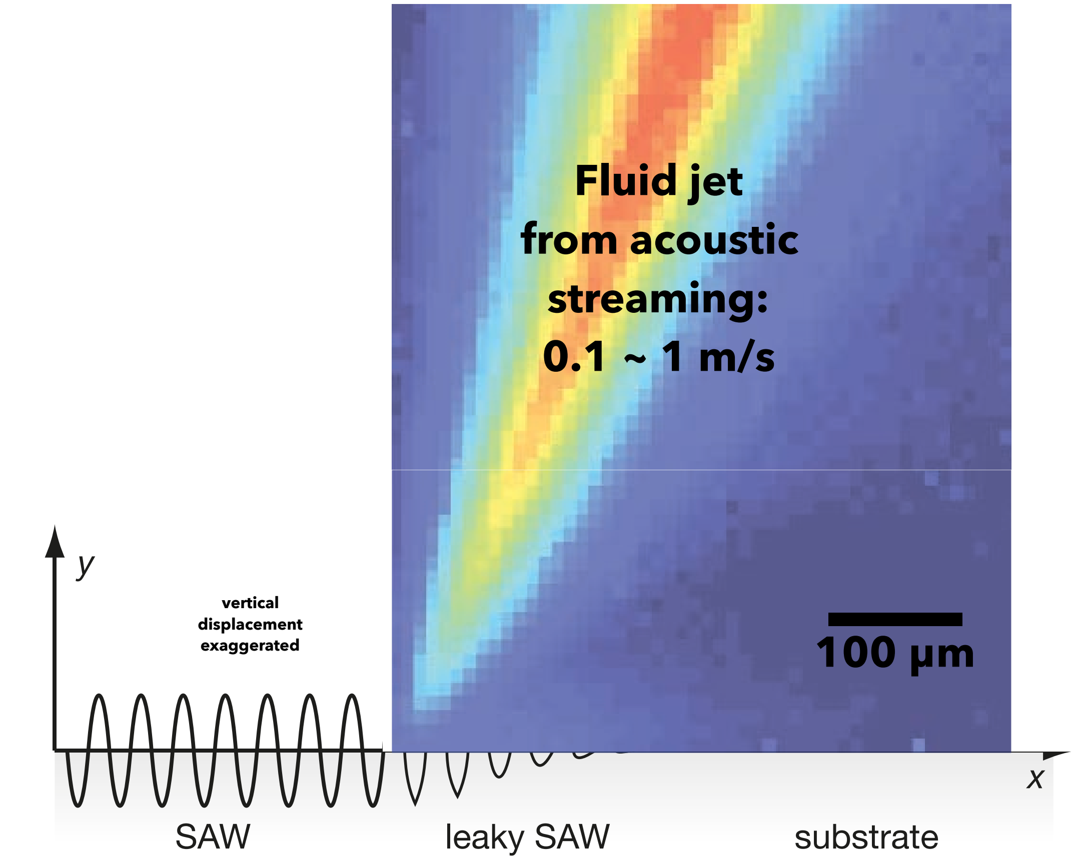
- We consider a "solid-state" method using (a) intense surface acoustic waves at ~100 MHz to drive acoustic streaming sufficient to drive flow in interelectrode gaps.
- Overcomes (b,c)* ion depletion adjacent the electrode during charging, particularly at high rates.
- Questions to address in this project:
  - **Why does it work?** ***ANALYSIS***
  - **Can it be made practical?** ***2 Ah PROTOTYPE***



* Results (b,c) are computed results of actual Li ion concentration in 50 μm interelectrode gap with separator.

Technical Accomplishment: SAW device design completed for integration into 2 Ah prismatic battery

- Successfully fabricated prototype 2 A-h pouch batteries incorporating SAW device



Frequency (MHz)	Attenuation in substrate (mm)	Attenuation in fluid (mm)	Position of max fluid vel. (mm)
19.7	2.4	120	~10
54.2	0.87	16	~6
122	0.39	3.1	~1
240	0.19	0.80	~0.5
490	0.097	0.19	~0.2
936	0.046	0.052	< 0.1

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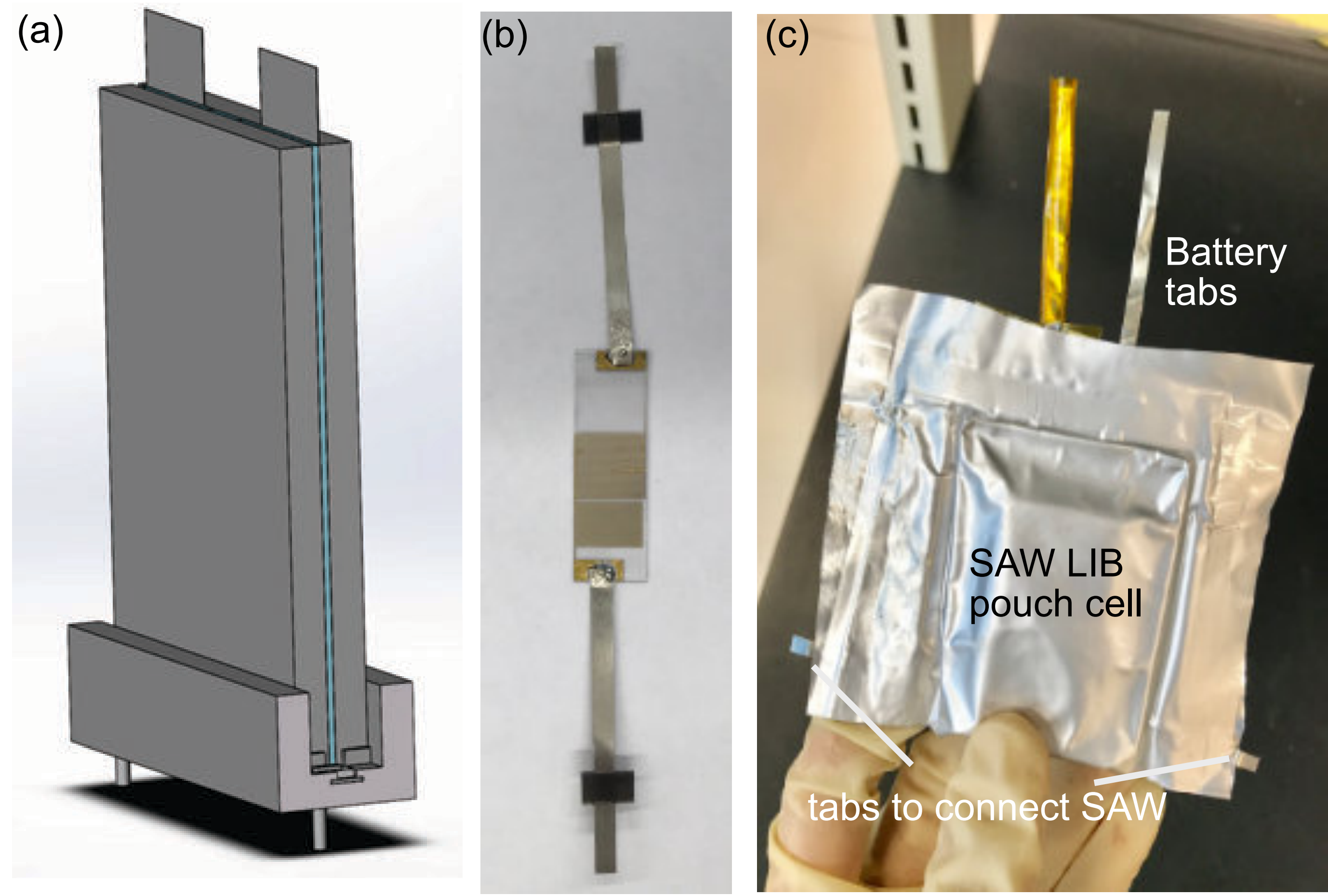
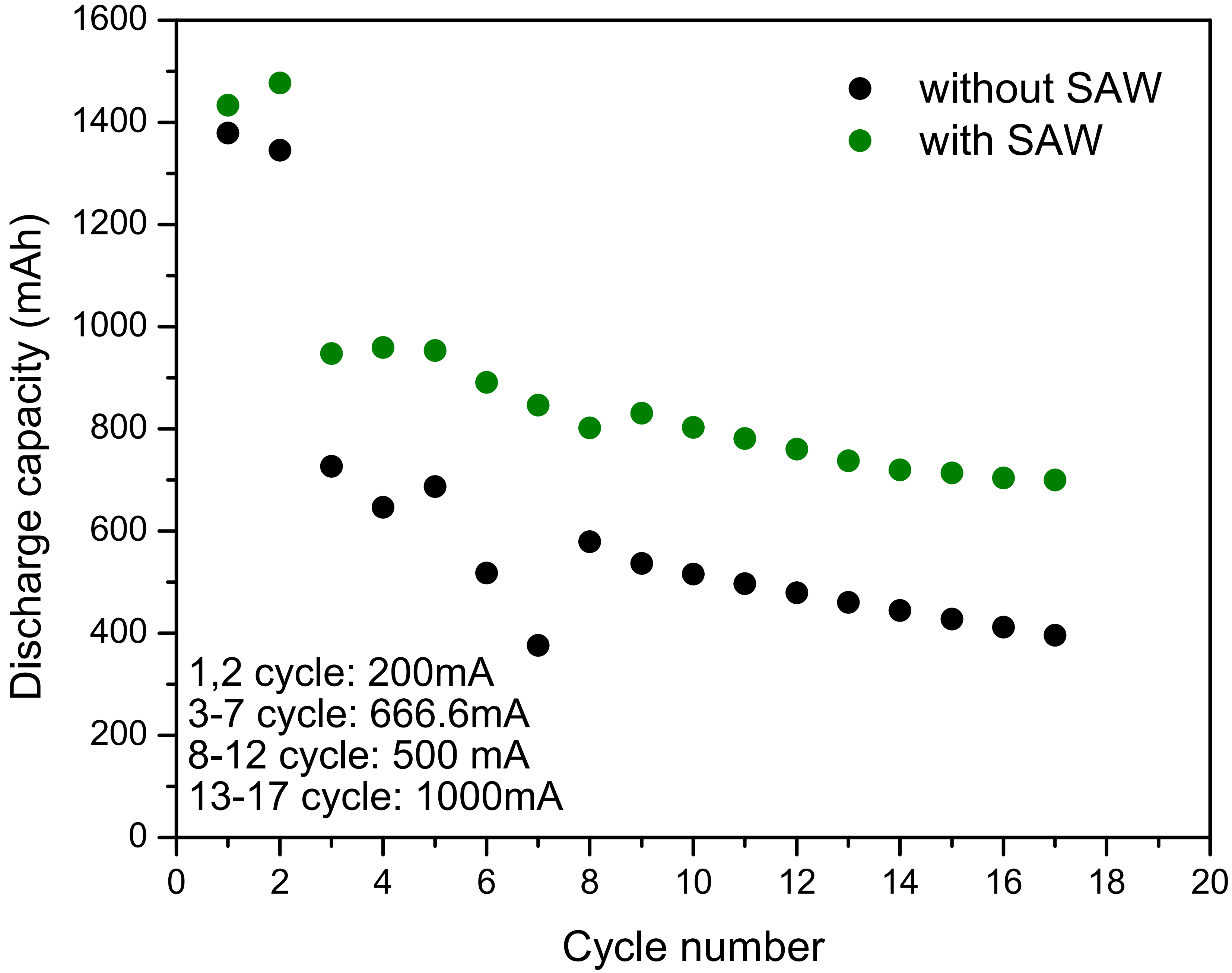
Technical Accomplishment: SAW device integration into 2 Ah prismatic battery

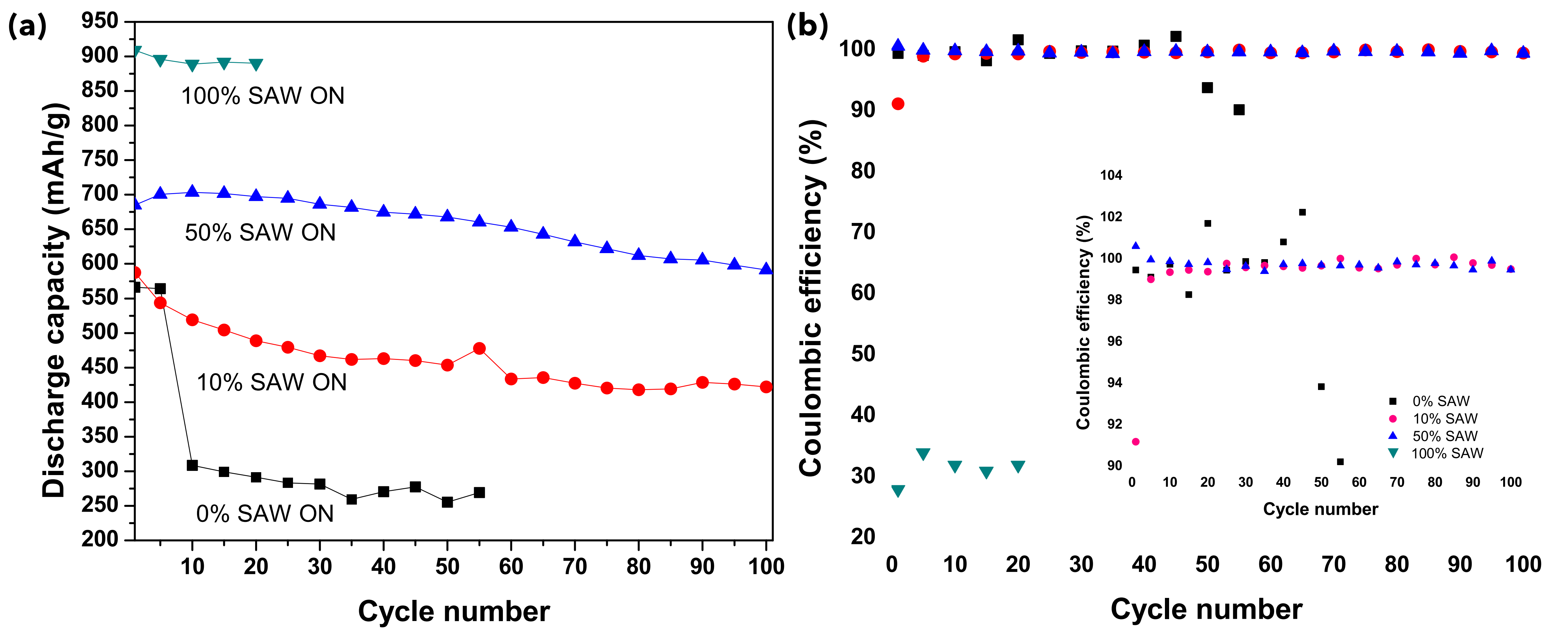
- Successfully fabricated prototype 2 A-h pouch batteries incorporating SAW device
  - (a) All-glass dummy battery for experimental visualization of interelectrode flow via micro/nano particle image velocimetry
  - (b) Example SAW device
  - (c) Completed prototype battery





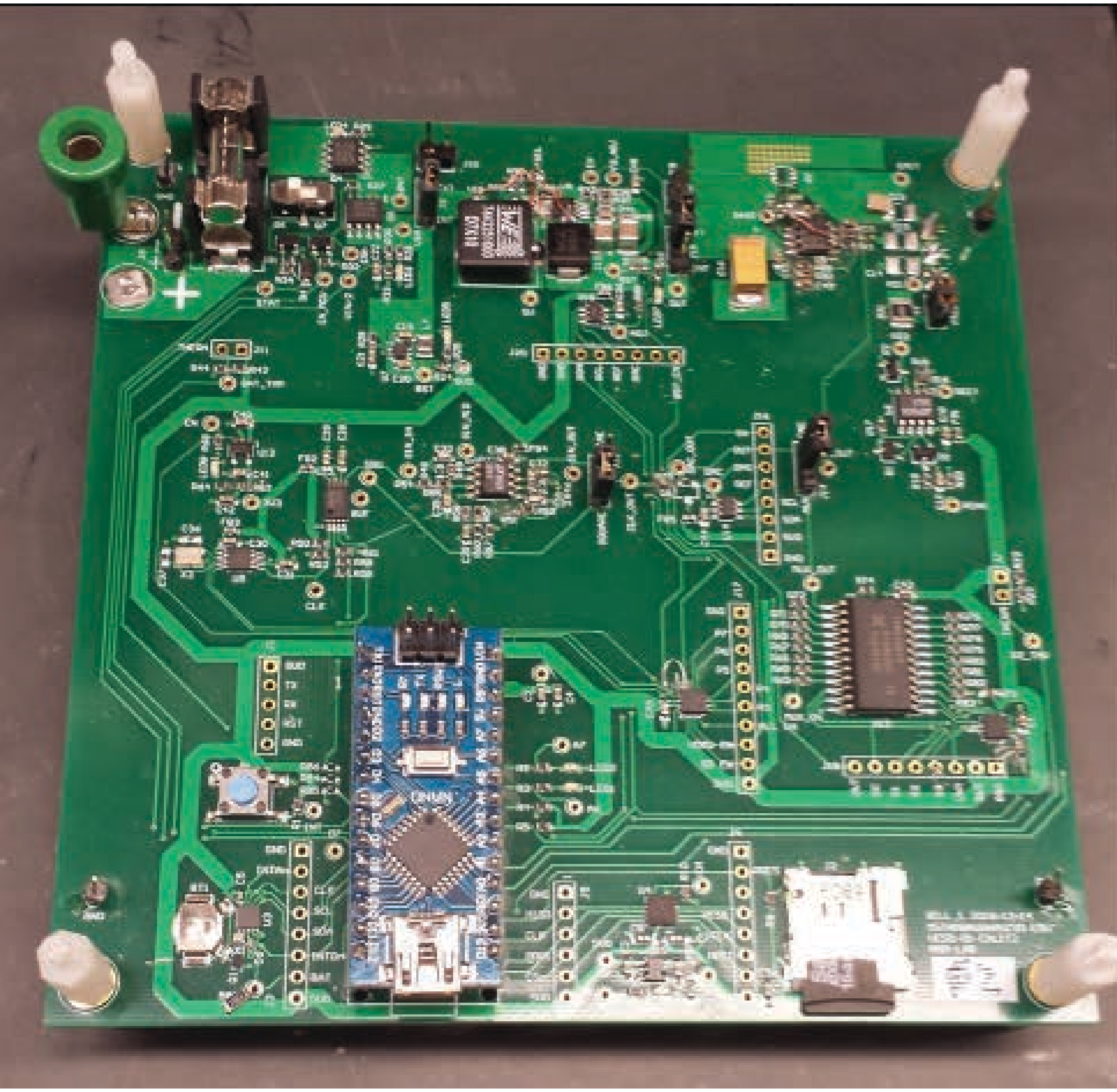
Technical Accomplishment: Cycling Prototype

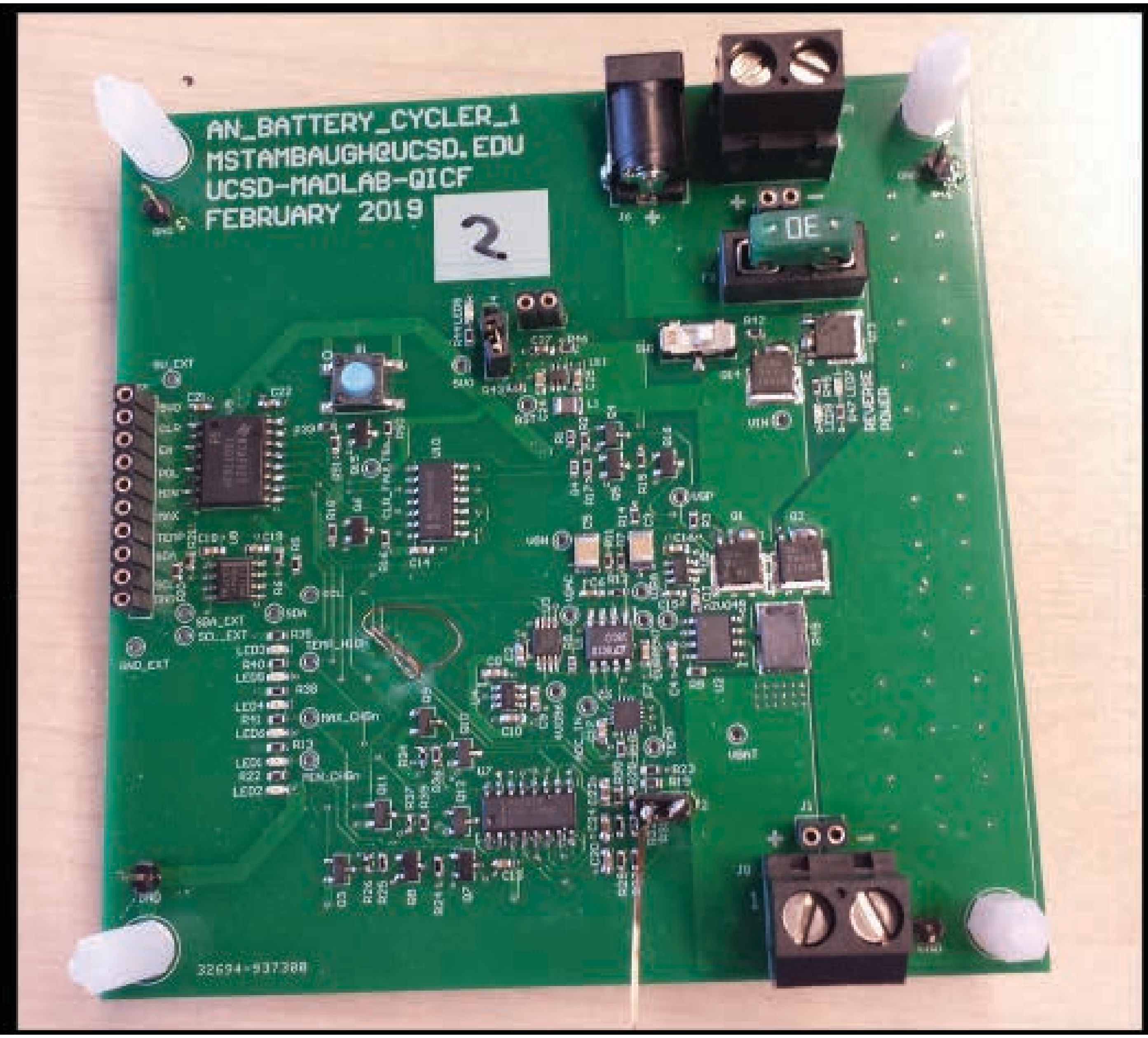
Prototype battery operating, shows increasing impact of using SAW in improving discharge capacity above 200 mA charge rates.



Technical Accomplishment: Effect of Duty Cycle for SAW

(a) modulating the SAW is effective and reduces the overall power required to operate it (from 200 mW max to as little as 20 mW at 10% duty cycle. (b) However, note the reduction in (c) Coulombic efficiency when the SAW runs at 100%. The reason is under study.

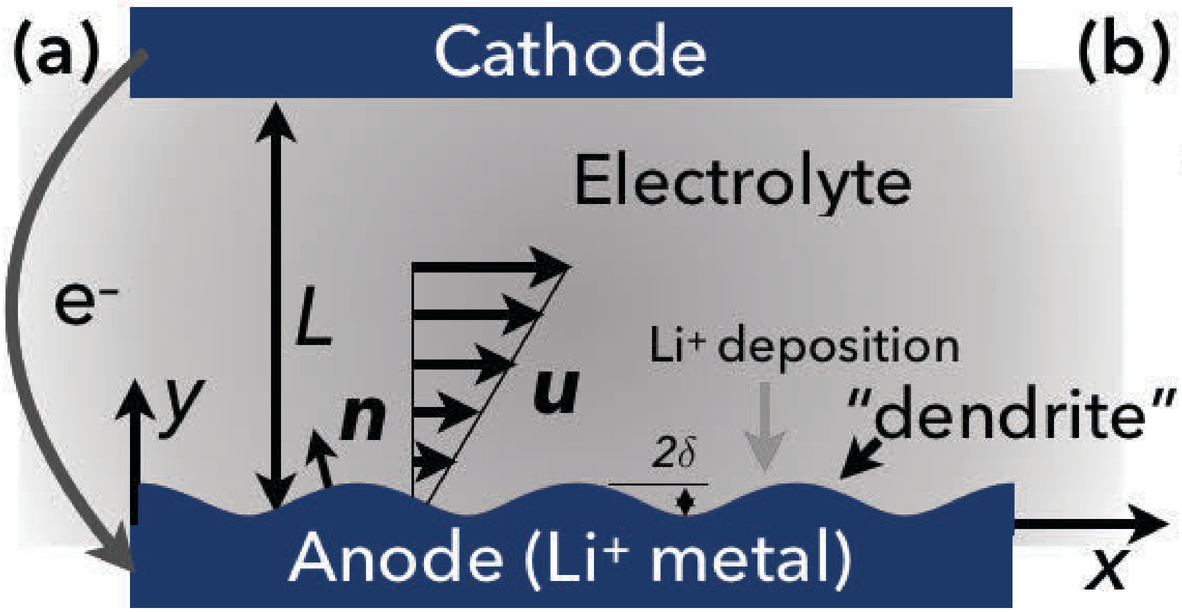




Technical Accomplishment: Prototype Driver System

We have devised a SAW device driver (left) and battery cycler capable of +/-30A (right) to demonstrate feasibility of concept. SAW driver is composed of components that can be reduced to 0.5 cm^3.

(a)



(b)

Li+ concentration

Diffusion coeff.

$$\frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = D\nabla^2 c$$

Fluid velocity

Convection-diffusion of Li ions

$$\iint_A c\, dA = A c_{\text{bulk}}$$

Electrolyte ion conc. is constant

$$c\,|_{t=0} = c_{\text{bulk}}$$

Ion conc. initial condition

Boundary condition along $y = 0 + \delta \cos kx$

$$-D\nabla^2 c \cdot \mathbf{n} = \alpha\sqrt{c\left[1 + \epsilon\left(1 + \cos kx\right)\right]}$$

Charge transfer coeff.

Perturbation

(c) No SAW…depletes Li+ ions and…

$$\frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = 0 = D\nabla^2 c$$

Convection-**diffusion** of Li ions

$$c = 0 - 1 + \epsilon\left(1 + \cos kx\right)$$

Boundary condition $y = 0 + \delta \cos kx$

…produces dendrites

$$i = -\frac{DF}{1 - t_+^0}\left(-D\frac{2c_{\text{bulk}}}{L} + \epsilon c_{\text{bulk}} k \cos kx\right)$$

Locally-enhanced Li+ deposition

An (a,b) analysis model has been devised with Prof Ofer Manor (Technion). Features: closed-form solution via intermediate matched asymptotics. (c) Without SAW, dendrites easily form from local ion depletion adjacent the presumably rough anode.

Technical Accomplishment: Analysis Model to Describe Phenomena (1/2)

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(d) With SAW…

Shear flow model

$$\mu \nabla^2 \mathbf{u} = \nabla p \quad \text{and} \quad \nabla \cdot \mathbf{u} = 0$$

Pressure

Dynamic viscosity

Fluid velocity along x

$$\frac{\partial u_x}{\partial x} = \beta \text{ as } y \to \infty \text{ and}$$

Tangent unit vector

$$\mathbf{u} \cdot \mathbf{n} = \mathbf{u} \cdot \mathbf{t} = 0 \text{ along } y = \delta \cos kx$$

Matched asymptotic expansion approach:

Inner region: $c = c_0 + c_1/\mathrm{Pe} + \cdots$
(ion convection and diffusion)

Outer region: $C = C_0 + C_1/\mathrm{Pe} + \cdots$
(ion convection only)

which will give the current flux $i = i_0 + i_1/\mathrm{Pe} + \cdots$

Length from acoustic source over which dendrite growth is suppressed:

Non-dimensionalize…

$$x \to \delta x \quad y \to \delta y \quad (u, v) \to u_c(u, v) \quad L \to \delta L \quad k \to k/\delta$$

…to produce…

$$\mathbf{u} \cdot \nabla c = D \nabla^2 c \to u \partial_x c + v \partial_y c = \frac{1}{\mathrm{Pe}} \left(\partial_{xx} c + \partial_{yy} c \right)$$

…and

with $\mathrm{Pe} = u_c \delta / D \gg 1$

$$\iint_A c\, dA = A c_{\text{bulk}} \to \frac{1}{A} \iint_A c\, dA = 1$$

$$c = -\epsilon \left(1 + \cos kx\right) \text{ along } y = 0.$$

…ion depletion & dendrites suppressed

$$x/\delta < \mathrm{Pe}^3 \left(3k\delta \frac{a_2}{a_1} \right)^{-3}$$

$a_1 \equiv 3^{1/3}(1-\epsilon)/\Gamma(1/3)$
$a_2 \equiv \sqrt{\pi}(3/2)^{1/3}/\Gamma(1/6)$

Technical Accomplishment: Analysis Model to Describe Phenomena (1/2)

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(d) With the flow induced by SAW acoustic streaming, there is **no turbulence**. However, the Peclet number becomes >>1, and the key is to ensure that the Pe is everywhere large to avoid ion depletion and dendrites.

Collaboration and Coordination with Other Institutions

- Prof. Ofer Manor, Dept Chemical Engineering, the Technion, Israel: analysis ideas and modeling of the acoustic streaming-enhanced electrochemistry in this system
- Profs. Ilenia Battiato and David Tartakovsky, Stanford Uni: analysis treatment for acoustic streaming at finite amplitudes and high frequencies as seen here
- Qualcomm Institute (UCSD; subcontractor): circuit design and prototyping help.

Remaining Challenges and Barriers

- A key challenge is achieving 6C charge rates for fully 500 cycles. We are seeing evidence of being able to achieve this goal, but have battery assembly and SAW integration problems to overcome to consistently deliver >4C charge rates to 500–1000 cycles for our 2 Ah prototypes.
- Another challenge is the design and assembly of the jellyroll battery. We have the SAW integration aspects solved for this configuration, but the key difficulty is entirely practical in fabricating 2 Ah jelly-roll batteries with consistent performance characteristics.

Proposed Future Research

- Upcoming is a transition from the prismatic pouch 2 Ah battery with SAW to the jelly-roll configuration, with the first such batteries to be produced late summer 2019.
- Our analysis is sufficiently advanced at this point to serve as an effective design tool, but relies upon simple assumptions regarding the anode surface morphology (wavy roughness). We intend to combine our COMSOL (computational) and extensions to our closed-form (asymptotics) analyses to deliver a model that predicts the necessary conditions to avoid ion depletion and dendrites with better veracity.
- If our progress remains on track, we may be able to achieve our stretch goal for this project in also examining the ability of our device to provide a safety function as an integrated pulse-echo sensor suitable for detecting dendrites, electrolyte loss, or other failure.
- We aim to obtain more data on the internal flow structure induced by the SAW device depending on operating conditions in dummy (transparent) prismatic and jelly-roll battery structures. This will support our analyses and provide guidance on design improvement.

Summary

- Have prototype prismatic batteries with integrated SAW devices that effectively recirculate the electrolyte within.
- The effect of using the SAW enables performance improvement of our prototype Li ion batteries.
- We have completed prototype driver circuits for the SAW device and battery cycler to +/- 30A appropriate for testing our batteries, demonstrating a key step towards a practical SAW LIB.
- We have a closed-form model for the electrochemical and acoustofluidic dynamics present in this system that suggests we must seek to maintain a large Peclet number to ensure rapid charging is possible. The phenomena does not occur due to “fluid turbulence” as previously believed.

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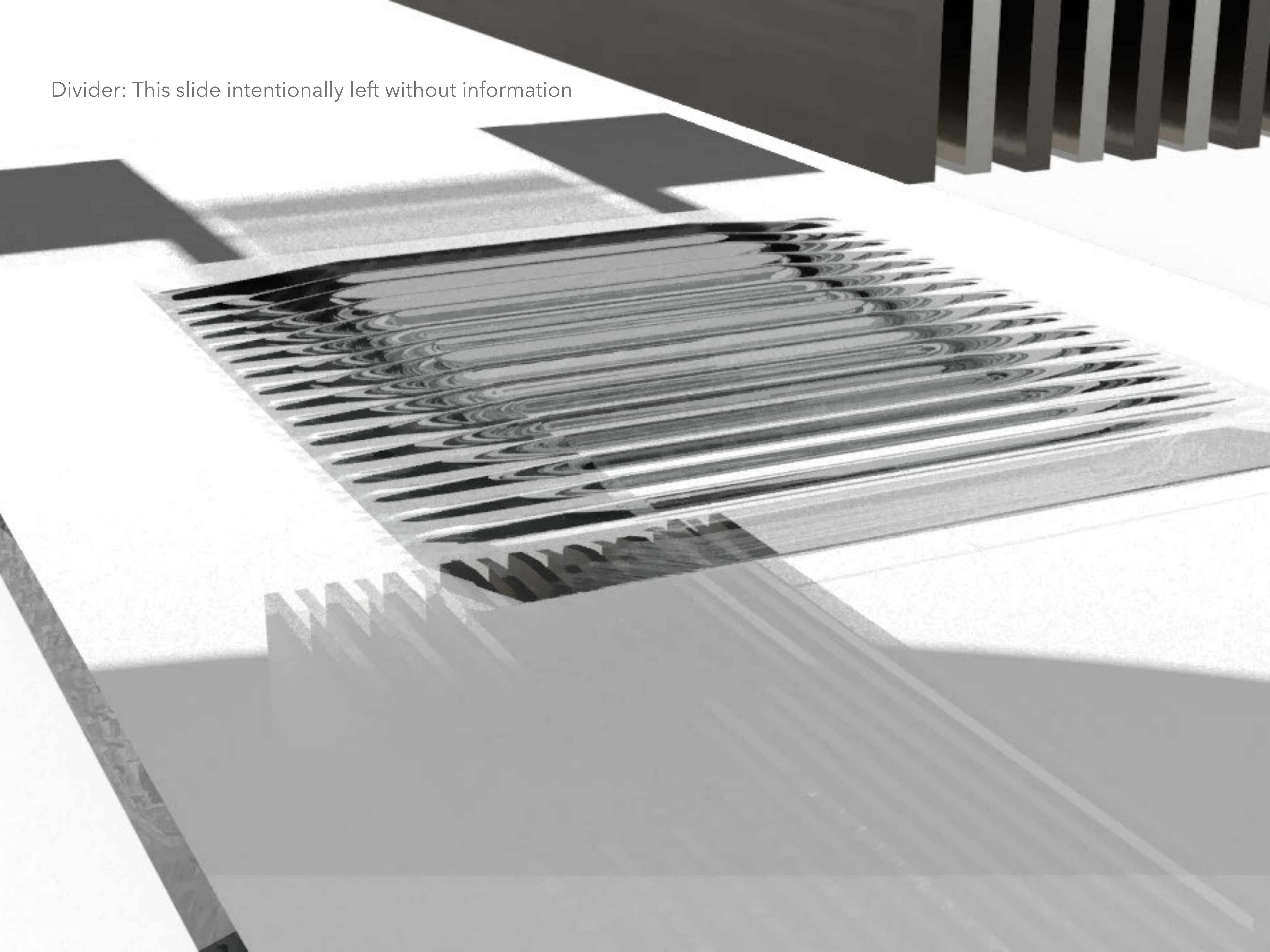
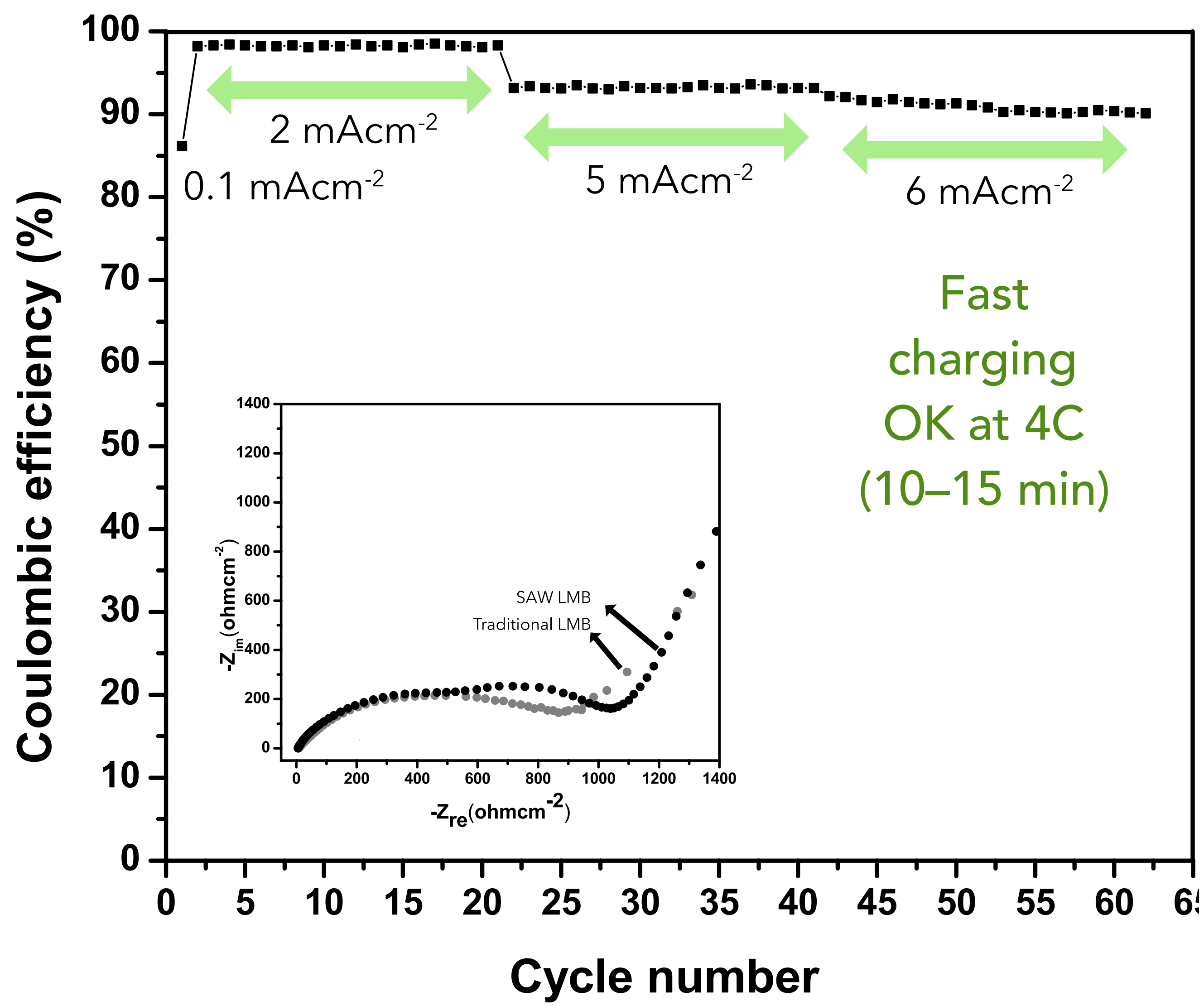
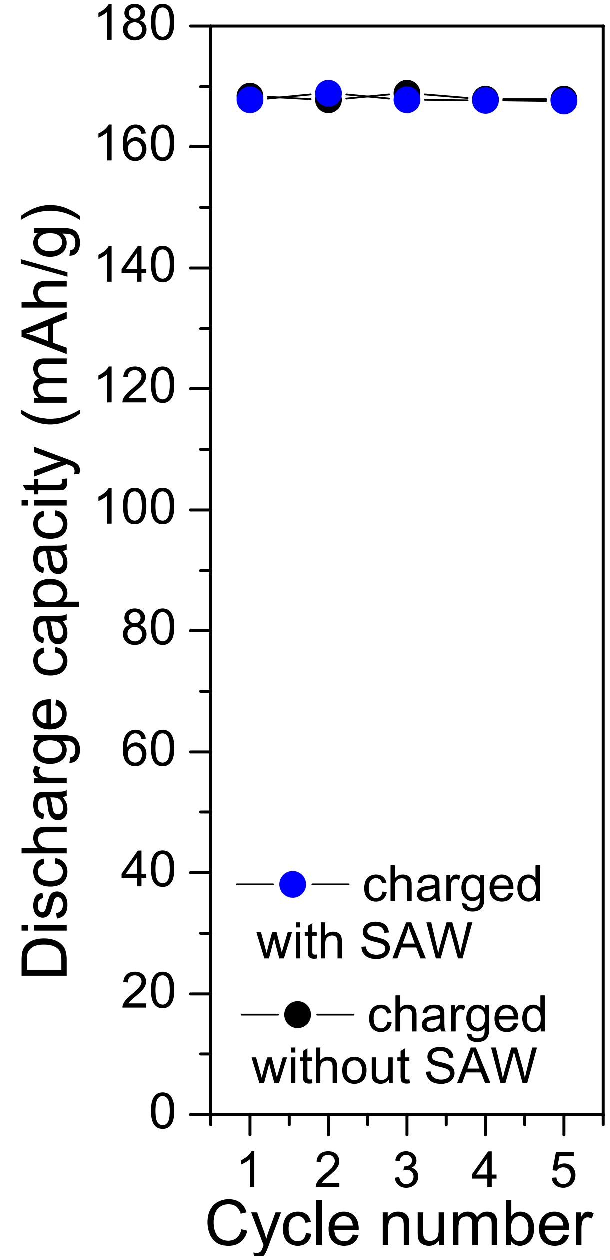




Extra slide: Li/ Cu cell

Switch to Cu cathode

Coulombic efficiency comparison at 2, 5, and 6 mA/cm^2 charge/discharge rates



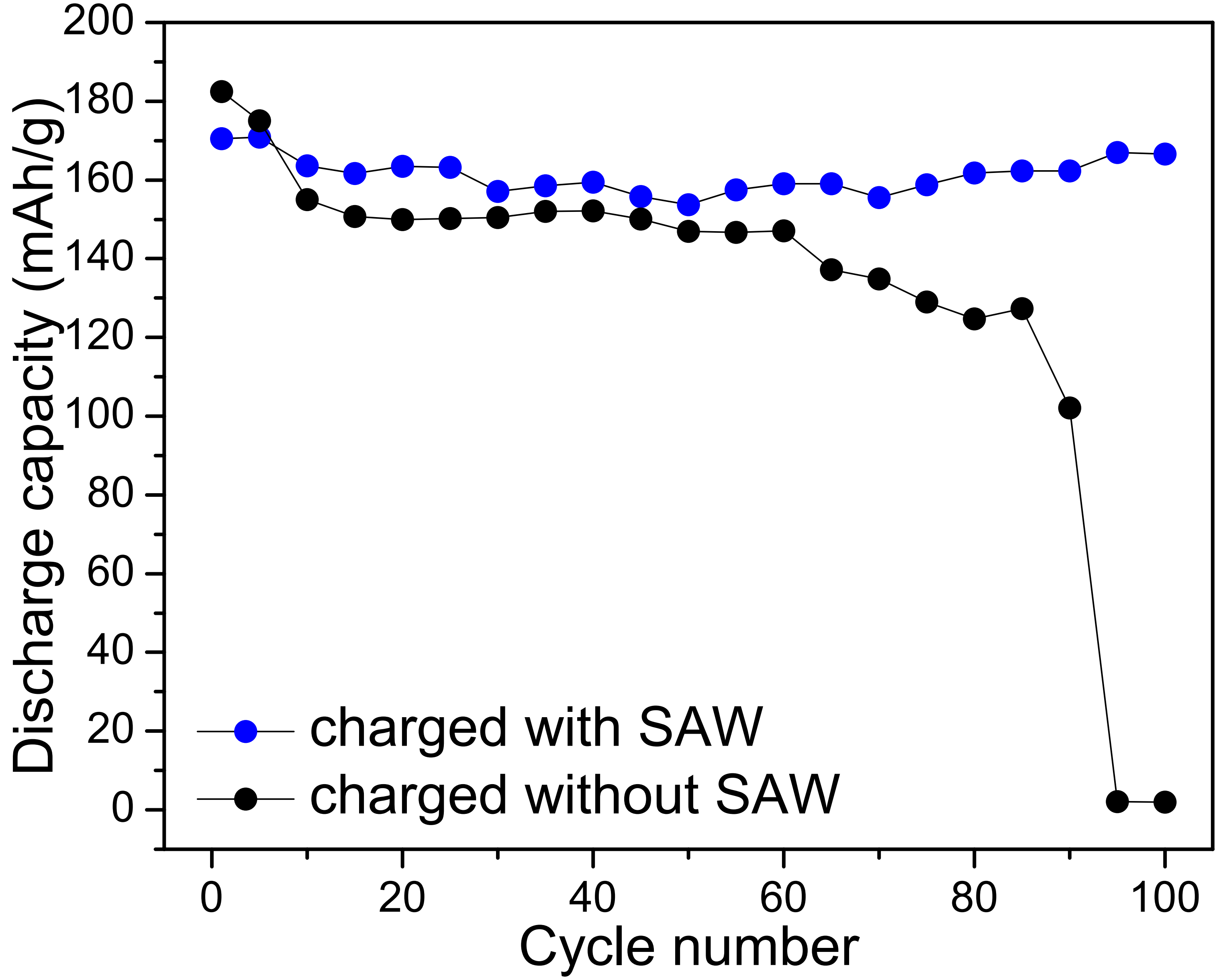
Extra slide: does SAW affect battery? *No.*

Charge/discharge cycling for 5 cycles…

(Cyclic voltammogrammetry)

Low charging rates (~0.1 C)

Cutoff voltages at 2.5 and 4.2 V (vs. Li/Li+)



Extra slide: Cycling performance?

Discharge capacity remains at or above 70 % theoretical for 500+ cycles at 3mA/cm^2 (2C)

cutoff voltages at 2.5 and 4.2 V (vs. Li/Li+)

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